Calibration of JGOFS/WOCE/OACES Carbon Measurements in the North Pacific Ocean

Robert M. Key November 15, 1999

1.0 Introduction

The first stated goal of the funded project, **Synthesis and interpretation of the NOAA/DOE Global CO2 Survey Data,** was to generate a unified carbon dataset composed primarily of results from the recent DOE/NOAA/NSF sponsored global survey. The planned procedure was that data from each cruise would be individually subjected to quality control (QC) procedures then the individual cruises would be examined to determine if there were any systematic cruise to cruise offsets. Where possible, systematic offsets would be determined using a procedure similar to that developed by Key (1998;

http://geoweb.princeton.edu/staff/Key/key.cross/crossover.html) for the Indian Ocean data. With this procedure one assumes that deep and bottom waters are in steady state with respect to the distribution of the various carbon system parameters. This assumption implies that the deep and bottom water measurements at the location(s) where different cruises intersect should have the same carbon distribution in density space. The crossover techniques proved valuable in the Indian Ocean where every cruise had one or more crossover points (Sabine *et al.*, 1998), however, in the North Pacific Ocean no zonal sections were carried out by U.S. investigators during the survey. Only 2 cruises, P16S17S and P17N provide crossover checks which compare different meridional sections. All other crossovers in the North Pacific only provide comparisons along an individual meridional section. Japanese scientists did sample the P1 and P2 zonal sections. When this data is available, crossover analysis between these two cruises and the meridional sections will provide either a check on or replacement of the results presented here.

This note briefly describes an alternate procedure to perform the cruise to cruise calibration. As with the crossover procedure, this technique assumes steady state for deep and bottom waters. Additionally, it is assumed that in these waters there is no chemical or biological process occurring which will markedly change the relationship between the carbon parameter in question and some subset of other routinely measured quantities including pressure, salinity, temperature, oxygen, nitrate, silicate, phosphate and quantities calculated from these measured values. When successful, the procedure should be able to detect systematic offsets which are greater than or equal to the measurement precision. Once any offsets have been found and corrected (whether by crossover analysis or regression analysis), the results of this calibration should produce a unified data set having an accuracy comparable to the measurement precision.

2.0 Procedure for TCO₂

2.1 Database Assembly

The database consists of all available high quality modern results. The vast majority of the cruise data were compiled from the recently completed WOCE (World Ocean Circulation Experi-

ment) survey. This data was supplemented by cruise data from the NOAA sponsored OACES program and a few other somewhat older cruises. In all cases the routine hydrographic data was collected from either the WOCE Hydrographic Office at SIO (http://whpo.ucsd.edu/), the NOAA data center, or directly from the individual chief scientist. Carbon data was collected either from CDIAC or from the principal investigator responsible for the measurements. Details of the various cruises is summarized at the GLODAP web site

(http://cdiac.esd.ornl.gov/cdiac/oceans/glodap/index.html). The Pacific Ocean cruises included in this particular study are summarized in Table 1.

TABLE 1. Modern Pacific Ocean Cruises with Carbon Measurements

rbon Measureine	ints
WOCE	Who
	WIIO
CGC91-1	NOAA
EQPAC Fall	NOAA
EQPAC Spring	NOAA
P10	WOCE
P13N	NOAA
P14N	WOCE
P14S15S	WOCE
P15N	WOCE/Canada
P16A17A	WOCE
P16C	WOCE
P16N	NOAA
P16S17S	WOCE
P17C	WOCE
P17E19S	WOCE
P17N	WOCE
P18	NOAA
P19	WOCE
P21	WOCE
P31	WOCE
P6	WOCE
P8S	WOCE/Japan
P9	WOCE/Japan
S4P	WOCE/Russia
	Cruise Designation CGC91-1 EQPAC Fall EQPAC Spring P10 P13N P14N P14S15S P15N P16A17A P16C P16N P16S17S P17C P17E19S P17N P18 P19 P21 P31 P6 P8S P9

2.2 Quality Control

Initial quality control for each cruise was performed by the principal investigator for each cruise. This procedure included checking the data for extreme outliers (which in most cases were totally eliminated) and frequently, in cases where Certified Reference Materials (CRM; Dickson, 1990, 1991, 1992; UNESCO, 1991) were available, adjusting the measurements so that the CRMs

measured at sea agreed with the certified value. These data were then submitted to CDIAC. Subsequent QC was carried out either by R. Key at Princeton and/or by A. Kozyr collaborating with the principal investigator and quality control flags conforming to WOCE guidelines (WHPO, 1991) were appended to each measured value. A listing of the samples which received a flag value other than "2" (good) can be found through a link on the GLODAP cruise summary table. This secondary QC was primarily done by looking for anomalous data in various property-property plots, but the investigation was always limited to "one cruise at time". Overall, the fraction of samples flagged as questionable (3) or bad (4) was less than 1% of the total.

2.3 Determine "Best" Fitting TCO₂ Equation

Subsequent to QC all of the cruise data listed in Table 1 were merged into a single uniform format database. Since this study was only concerned with calibration for the North Pacific (where no crossovers exist), a subset including only measurements collected north of the equator was taken. To be consistent with the Indian Ocean crossover study and to minimize the potential error in the steady state assumption, the data was further subsetted to include only samples collected at pressures greater than 2500 dB. The resulting database included just over 1800 samples and was limited to those cruises in Table 1 which are in **bold** text.

Based on the work of Brewer *et al.* (1997), Wallace (1995), Sabine *et al.* (1999) and others, a simple linear equation with intercept, which included only commonly measured parameters as the independent variables, was sought. The search was thorough, but not exhaustive. An initial guess of important terms was made, then terms were added or eliminated based on examination of the residuals relative to both included and excluded terms, and upon the statistical significance of the included terms, respectively. During the search, additional terms were not retained in the function unless the residual error of the fit improved considerably (a subjective judgment based on error decrease relative to change in the degrees of freedom). The resulting equation was of the form shown in Equation 1 and summary statistics for this regression are summarized in Table 2:

$$TCO_2 = a + bNO_3 + cAOU + dSi + eSalt + fSalt^2$$
 (EQ 1)

TABLE 2. Summary Statistics for Initial Fit

	•			
Coefficient	Value	Std. Error	t Value	Pr (> t)
Intercept	-5108803	461715	-11.1	0.0000
NO_3	1.29	0.20	6.5	0.0000
AOU	.969	.025	38.2	0.0000
Si	0.122	0.015	8.1	0.0000
Salt	294430	26641	11.0	0.0000
Salt ²	-4240	384	-11.0	0.0000

Residual standard error: 4.143 on 1800 degrees of freedom

Multiple R-Squared: 0.9494

F-statistic: 6752 on 5 and 1800 degrees of freedom, the p-value is 0

The residuals from this fit showed a slight positive correlation with longitude, however, this appeared to be primarily due to the fact that the westernmost cruise, P8S, was anomalously low with respect to the other cruises. The last term in Equation 1, Salt² could be eliminated by writing the equation in terms of total carbon normalized to a constant salinity.

2.4 Identify Anomalous Cruises and Refit

Figure 1 shows a boxplot summarizing the residuals from the fit using Equation 1 and all of the North Pacific data. The dotted lines indicate one standard deviation for the fit. The length of each bar is indicative of the scatter for the cruise and the location shows how the various cruise results scatter relative to the fit. In this case four cruises, CGC91-1, P8S, P9 and P16C appear somewhat anomalous. Additionally worth noting is that the scatter in the residuals for P17N is

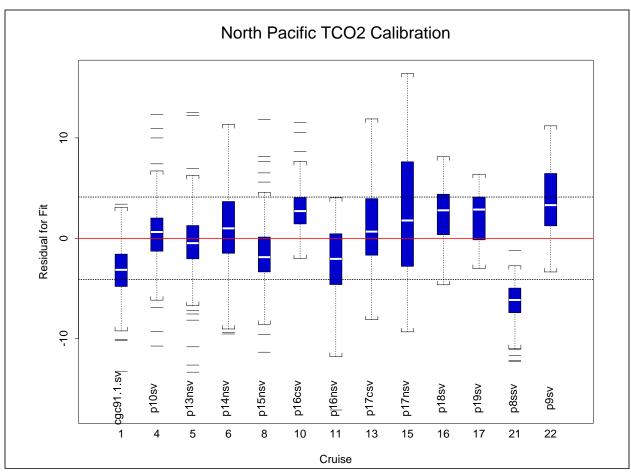


Figure 1: Residual summary including all North Pacific data

significantly larger than for the other cruises. Before preceding further, the residuals from this initial fit were examined in detail to try to identify if there were underlying reasons for the apparent misfit to the 4 mentioned cruises. First all of the residuals were plotted against latitude, longitude, pressure, salinity, silicate, nitrate, and oxygen looking for potentially important parameters, but no trends were found. Next the residuals were binned into 20° width latitude bands and examined as a function of longitude (Figure 2). Since most of the cruises were meridional sections, this technique essentially grouped the residuals by cruise with some merging. The only thing worth noting was that P8S, P9 and CGC91-1 still appeared anomalous. Talley and Joyce (1992) clearly demonstrated a strong (and anomalous relative to the rest of the Pacific) silicate source in the northeastern Pacific. Since P17N showed deviation in its residuals than the other cruises and P17N is in the same area identified as the silicate source, the possibility of a connection between the two was investigated. Here it was assumed that if the large spread in the P17N residuals was due to an anomalous silicate source then residuals for the region should show a pattern when plotted against pressure. To investigate the residuals were binned into 30° wide longitude bands and

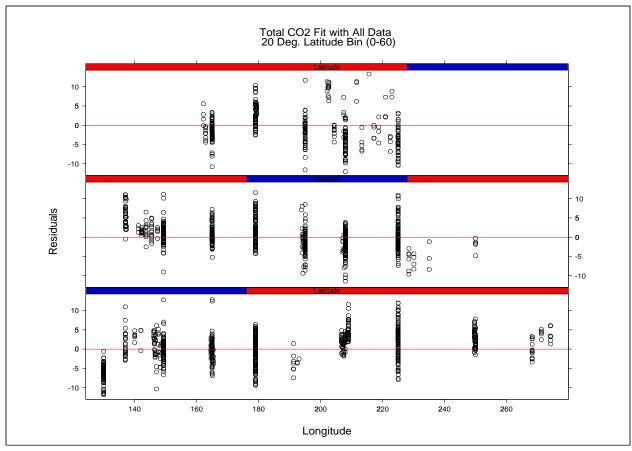


Figure 2: Residuals from Equation 1 using all North Pacific data binned into 20° latitude boxes and plotted against longitude. Other than the P8S and P9 residuals which are anomalously low and high, respectively, no convincing trends are seen.

20° wide latitude bands. The binned residuals were then plotted against pressure. Figure 3 shows the result which does not support the assumption. Finally, in a last attempt to rationally resolve the apparently anomalous cruises, the residuals were binned by cruise and plotted as a function of latitude. Because of the way the cruises were run, this is essentially the same as plotting as a function of time along the cruise line (either forward or reverse). Additionally, a simple linear regression was calculated for each set of residuals. For cases having $r^2 \ge 0.1$ and at least 30 measurements, the regression line was added to the plot. For all cases the largest r² was approximately 0.3. None of these correlations are particularly impressive, however it is interesting that each of the 4 cruises initially identified as anomalous show a trend. Also worth noting is the fact that all cruises showing a trend in the western Pacific (P8S, P9 and P14N) are positively correlated while those in the eastern Pacific are negatively correlated (CGC91-1, P16C, P17C and P18). Since latitude is approximately equivalent to time, these correlations are what one would expect to see if the analytical equipment response was drifting. Detecting and correcting for instrument drift was one of the primary reasons that Certified Reference Materials (CRM) were developed for TCO₂. On all of the cruises showing a trend with time (with the possible exception of P8S), CRMs were measured for TCO₂. However, records indicate that only CGC91-1 and P18 had the TCO₂ results corrected to the CRM measurements. Current information is insufficient to determine whether the trends seen in Figure 3 were due to instrument drift, a real pattern in the TCO_2 distribution which was not captured by the fitting equation, or just chance.

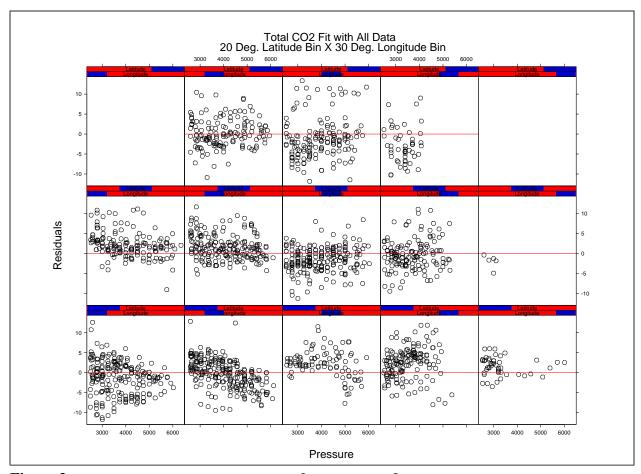


Figure 3: Residuals from Equation 1 binned into 30° longitude by 20° latitude boxes and plotted against sampling pressure. Left to right is west to east and bottom to top is south to north. If the large spread in the residuals for cruise P17N were due to anomalous silicate input, then one could reasonably expect to see a pattern in the residuals for the northeastern Pacific. No such trend is seen here.

Since we are seeking a calibration which is as near the measurement precision as possible, results from these four cruises were *assumed* to be offset and were eliminated. Data from the remaining 9 cruises were refit using the same function (Equation 1). The results from this refit are

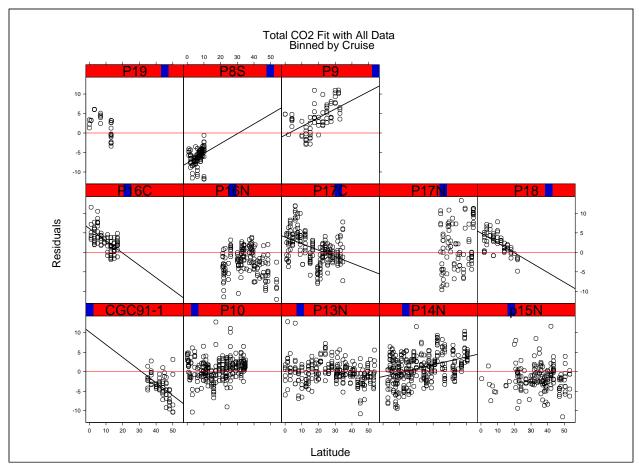


Figure 4: Residuals from the fit using Equation 1 and all North Pacific data binned by cruise and plotted against latitude. Since the cruises were generally run as meridional sections, plotting against latitude is approximately equivalent to plotting against time. For cruises with more than 30 data points and $r^2 \ge 0.1$ a simple linear regression line indicating the trend is added. Even the strongest correlation had $r^2 \le 0.4$. This type trend could be indicative of instrumental drift, failure of the fitting function or chance.

summarized in Table 3 and Figure 5. No pattern was found when the residuals from the final fit

TABLE 3. Summary statistics for final TCO₂ fit

Coefficient	Value	Std. Error	t Value	Pr (> t)
Intercept	-6705967	47120	-14.2	0.0000
NO_3	0.65	0.20	3.3	0.0009
AOU	1.011	0.027	38.1	0.0000
Si	0.107	0.015	7.1	0.0000
Salt	386586	27190	14.2	0.0000
Salt ²	-5570	392	-14.2	0.0000

Residual standard error: 3.851 on 1512 degrees of freedom

Multiple R-Squared: 0.9577

F-statistic: 6843 on 5 and 1512 degrees of freedom, the p-value is 0

were plotted against any measured parameter.

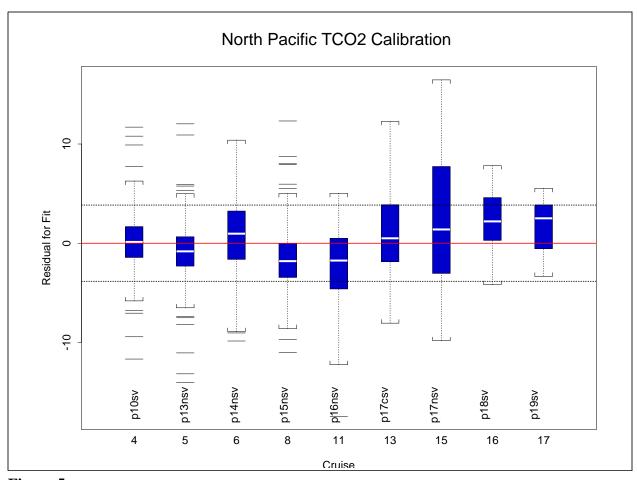


Figure 5: Residual summary for final fit of North Pacific TCO₂ data.

2.5 Estimate Appropriate Outlier Corrections

The coefficients listed in Table 3 were used with the measured nitrate, aou, silicate and salinity values from the four cruises which were not included in the fit to estimate TCO_2 values. The mean difference between the measured and estimated TCO_2 is taken to be the calibration factor for each cruise. These offset values are summarized in Table 4.

TABLE 4. TCO₂ Offset Summary for North Pacific

Cruise	Mean Difference EstMeas.	Standard Deviation	Standard Deviation of Mean
CGC91-1	3.7	3.2	0.4
P8S	7.2	2.0	0.2
P9	-3.3	3.8	0.5
P16C	-3.0	2.8	0.3

3.0 Procedure for Alkalinity

The same procedure used for TCO_2 was next applied to the North Pacific alkalinity data. The best fitting function was of the form shown in Equation 2 where PO^* is defined as $PO^* = O_2 + 170PO_4$ and Θ is the potential temperature.

Alkalinity =
$$a$$
Salt+ b Si+ c PO+ $d\Theta$ (EQ 2)

The residual plot and statistics for this equation using all of the North Pacific data are summarized in Table 5 and Figure 6, respectively.

TABLE 5. Summary Statistics for alkalinity fit with all data

Coefficient	Value	Std. Error	t Value	Pr(> t)
Intercept	-21439	1019	-21.0	0.0000
Salt	681	29	23.2	0.0000
Silicate	0.79	0.023	34.6	0.0000
PO	0.096	0.024	4.0	0.0001
Theta	57.7	2.1	27.5	0.0000

Residual standard error: 6.028 on 1111 degrees of freedom

Multiple R-Squared: 0.6891

F-statistic: 615.7 on 4 and 1111 degrees of freedom, the p-value is 0

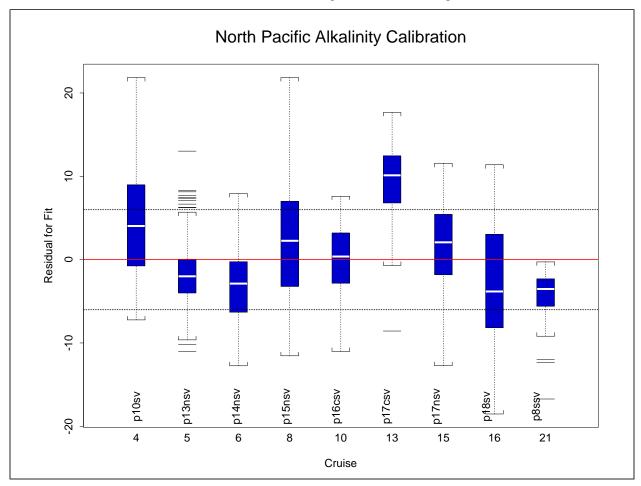


Figure 6: Residual plot for all North Pacific alkalinity data.

Based on this initial fit, it was assumed that the alkalinity data from cruises P8S and P17C needed calibration. Data from these cruises were excluded and the regression was recalculated.

The result from this second regression were used to estimate the average offset for the two eliminated cruises. The residual plot and regression statistics for the final fit are summarized in Table 6 and Figure 7. The residuals from the final fit were plotted against all individual hydrographic

TABLE 6. Final Regression Statistics for alkalinity fit

Coefficient	Value	Std. Error	t Value	Pr(> t)
Intercept	-20075	1004	-20.0	0.0000
Salt	641	29	22.2	0.0000
Silicate	0.74	0.024	31.7	0.0000
PO	0.12	0.024	4.9	0.0000
Theta	58.0	2.0	28.3	0.0000

Residual standard error: 5.849 on 1057 degrees of freedom

Multiple R-Squared: 0.7018

F-statistic: 621.9 on 4 and 1057 degrees of freedom, the p-value is 0

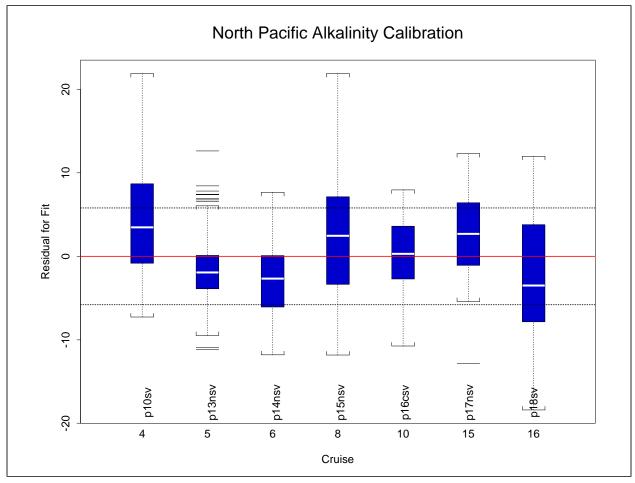


Figure 7: Residual plot for North Pacific cruises which were assumed to need no calibration.

parameters, however no further correlations were implied. Additionally, the coefficients used showed no cross correlation with the exception of the salinity coefficient and the intercept term which were highly correlated. This correlation was expected and indicates that a similar regression would have been obtained had the salinity normalized alkalinity been used for the dependent variable.

The coefficients given in Table 6 were then used to calculate estimated alkalinity values for cruises P8S and P17C. The mean difference between the estimated and measured alkalinity values is summarized in Table 7.

TABLE 7. Alkalinity offset summary for the North Pacific

Cruise	Mean Difference EstMeas.	Standard Deviation	Standard Deviation of Mean
P8S	5.4	4.0	0.73
P17C	-9.7	5.8	1.16

4.0 Summary

This exercise has demonstrated that it is possible to derive a linear equation in commonly measured parameters which fits high quality measured TCO₂ and alkalinity data to a precision *similar* to the measurement precision. For the North Pacific the TCO₂ fit was significantly better than the alkalinity fit. This was expected given the known difficulties with alkalinity determinations. *If* one accepts the assumptions stated at the beginning of this report, then the technique does provide an alternate method to detect cruises which are systematically offset for some reason and those offsets can be corrected. On the other hand, one can imagine many reasons why this technique might give erroneous results. Significantly more confidence could be placed in the regression method once the crossover analysis (including the Japanese data) has been completed.

5.0 References

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Addendum to OTL Tech. Report 99-3: Reestimate with Cruise P2

Robert M. Key March 3, 2000

Final calibration of the North Pacific WOCE carbon data sets based solely on the various crossover techniques is more problematic than for the South Pacific. P2 (EXPOCODE 49K6KY9401) was the only zonal section in the entire basin with high quality carbon measurements. This fact severely limits the number of potential crossovers and consequently places extreme statistical weight on the data from that cruise. In this case the uncertainty of the crossover analyses is increased by the fact that the scientists responsible for that cruise did not participate in the early DOE sponsored CO2 training sessions thus introducing at least one additional degree of freedom into the comparison. On the other hand, the independence of the P2 data provides an opportunity to identify bias that might otherwise be missed. These factors prompted us to seek a totally independent calibration method to supplement the crossover results.

Brewer, et al. (1995) and subsequently others (Wallace, 1995; Sabine et al., 1999), have clearly demonstrated that both TCO2 and TA concentrations in deep and bottom waters (i.e. those that can reasonably be assumed to be at steady state) can be described quite accurately by multiparameter linear functions using commonly measured hydrographic quantities for the independent parameters. The extent over which any such function is applicable depends on the number of water masses present and the uniformity of chemical and biological processes which have effected the carbon concentration in each water mass. Relative to the rest of the ocean, the deep North Pacific is extremely uniform in almost all properties and is therefore an ideal region with respect to Brewer's finding.

The variation in the linear regression procedure for this study was to investigate the residuals for evidence of systematic differences between the various cruises. Details of the procedure can be found in Key (1999). For TCO2 the best functional form found for all of the North Pacific data is given by EQ1 where TCO2, NO3, AOU, and Si are the measured total inorganic carbon, the inorganic nitrate, the apparent oxygen utilization and the inorganic silicate concentrations in umol/kg and Salt is the salinity relative to the practical salinity scale. The form of EQ 1 is a bit unusual in that it contains both a salinity and a salinity ² term. The square term was included because without it the residuals were correlated with salinity. Similarly, a salinity dependence exists in the residuals if carbon normalized to a constant salinity is substituted for carbon concentration. Applied to all the North Pacific data the regression had a residual standard error of 4.8 umol/kg on 2082 degrees of freedom and a multiple R² of 0.93. The residuals from this fit were binned by cruise and examined. Two cruises, CGC91-1 and P8S, had average residual values greater in magnitude than the standard error for the fit. (-4.9 umol/kg and -6.5 umol/kg, respectively).

$$TCO_2 = a + bNO_3 + cAOU + dSi + eSalt + fSalt^2$$
 (EQ 1)

The P8S TCO2 data were deemed anomalous. The regression was recalculated excluding these data with a resulting standard error of 4.6 umol/kg on 1978 degrees of freedom and multiple R² of 0.94. The coefficients for this fit and the average residual segregated by cruise are given in

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Table 1 and Table 2, respectively. The last row of Table 2 shows the equivalent values between

TABLE 1. Equation 1 Coefficients

Coefficient	Value	Error
a	-6530293	485401
b	0.82	0.18
c	0.91	.03
d	0.14	.02
e	376554	28009
f	-5426	404

TABLE 2. Residual Summary for final TCO2 Regression

Cruise	Residual Average	Residual Standard Deviation	n	Residual Standard Deviation of mean
CGC91-1	-4.3	3.6	63	0.4
P10	-0.7	2.8	255	0.2
P13N	-1.7	2.9	258	0.2
P14N	04	3.7	325	0.2
P15N	-2.3	3.6	175	0.3
P16C	2.5	2.8	85	0.3
P16N	-3.1	4.1	166	0.3
P17C	0.5	4.1	195	0.3
P17N	2.6	6.7	96	0.7
P18	2.8	2.6	58	0.3
P19	3.1	3.4	28	0.6
P9S	1.9	3.7	73	0.4
P2	4.3	5.4	207	0.4
P8S ^a	-8.4	2.2	104	0.2

a. estimated from final regression refit

estimated values using the coefficients from Table 1 in Equation 1. In the second column of Table 2 the average values < 0 indicate that data from the cruise would need to be increased to match results predicted by the final fit. This result indicates that an upward adjustment of ~8.4 umol/kg is required for P8S and an upward and downward adjustment of ~4 umol/kg is suggested for cruises CGC91-1 and P2, respectively. The other cruise residual averages are too close to the replicate measurement precision to warrant adjustment. Applying Equation 1 to the GEOSECS hydrography implies that the GEOSECS TCO2 data in the North Pacific require a downward adjustment of 20.7+/- 14.8(0.71 for average; n=432) umol/kg. Approximately 15 umol/kg difference was accounted for previously by a programming error (T. Takahashi reference to Broecker and Takahashi, 1978, in Broecker, Spencer and Craig, 1982).

An analogous procedure was carried out for the North Pacific alkalinity data. In this case the best fitting function found was Equation 2. After the initial regression cruises P2, P8S and P17C were deemed anomalous. No significant correlation was found between the residuals and any other hydrographic parameter or between location other than explained by cruise. Measurements from these three cruises were eliminated and the regression recalculated. Table 3 gives the regression coefficients for this final fit and Table 4 summarizes the residuals. Using Equation 2, the coefficients from Table 3 and the measured hydrographic data, alkalinity values were esti-

mated for the 3 eliminated cruises. The average difference between the estimated and measured alkalinities for these 3 cruises represent the best estimate of the adjustment necessary to make these results comparable to the remaining cruises and are summarized in Table 5.

Alkalinity =
$$a$$
Salt+ b Si+ c PO+ $d\Theta + e$ (EQ 2)

TABLE 3. Coefficients for Equation 2

Coefficient	Value	Error
a	629	30
b	0.74	0.02
c	0.12	0.02
d	57	2
e	-19657	1073

TABLE 4. Residual Summary for Final Alkalinity Fit

Cruise	Residual Average	Residual Standard Deviation	n	Residual Standard Deviation
		Deviation		of mean
P10	4.5	6.7	215	0.5
P13N	-1.9	3.6	253	0.2
P14N	-3.0	3.8	284	0.2
P15N	2.4	6.6	121	0.6
P16C	0.2	4.3	50	0.6
P17N	2.6	5.4	71	0.6
P18	-2.3	7.1	58	0.9

TABLE 5. Implied Alkalinity Adjustments

Cruise	Residual Average	Residual Standard Deviation	n	Residual Standard Deviation of mean
P2	17.1	4.2	186	0.3
P8S	5.6	4.0	29	0.7
P17C	-8.7	7.6	26	1.5

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